Computing Grand Challenge Symposium

Toward Petascale Atomistic Simulations with Quantum-Level Accuracy

John A. Moriarty

H Division, Physical Sciences Directorate

February 20, 2008

Co- Investigators: Lin H. Yang, Randy Q. Hood

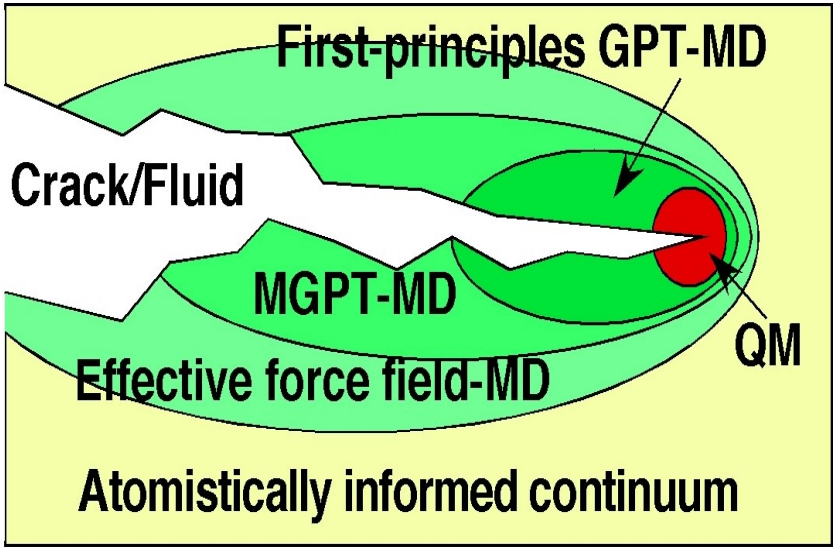
Collaborations: USC (Vashishta *et al*.), Harvard (Kaxiras *et al*.), LANL (Voter *et al*.)

This work was performed under the auspices of the U. S. Department of Energy by the Lawrence Livermore National Laboratory under contract No. DE-AC52-07NA27344

Motivation: recently funded SciDAC-2 project on stress corrosion cracking (SCC)

- Corrosion is complex technical problem (in e.g., advanced power generation) with annual economic impact equal to 3% of GNP
- USC, Harvard, CSUN, Purdue, LANL, LLNL collaboration
- *5-year goal*: develop hierarchical petascale simulation framework to address SCC from first principles
- *LLNL role*: develop new capability to perform ultra-scale atomistic simulations (10^6–10^{12} atoms) with quantum-mechanical accuracy

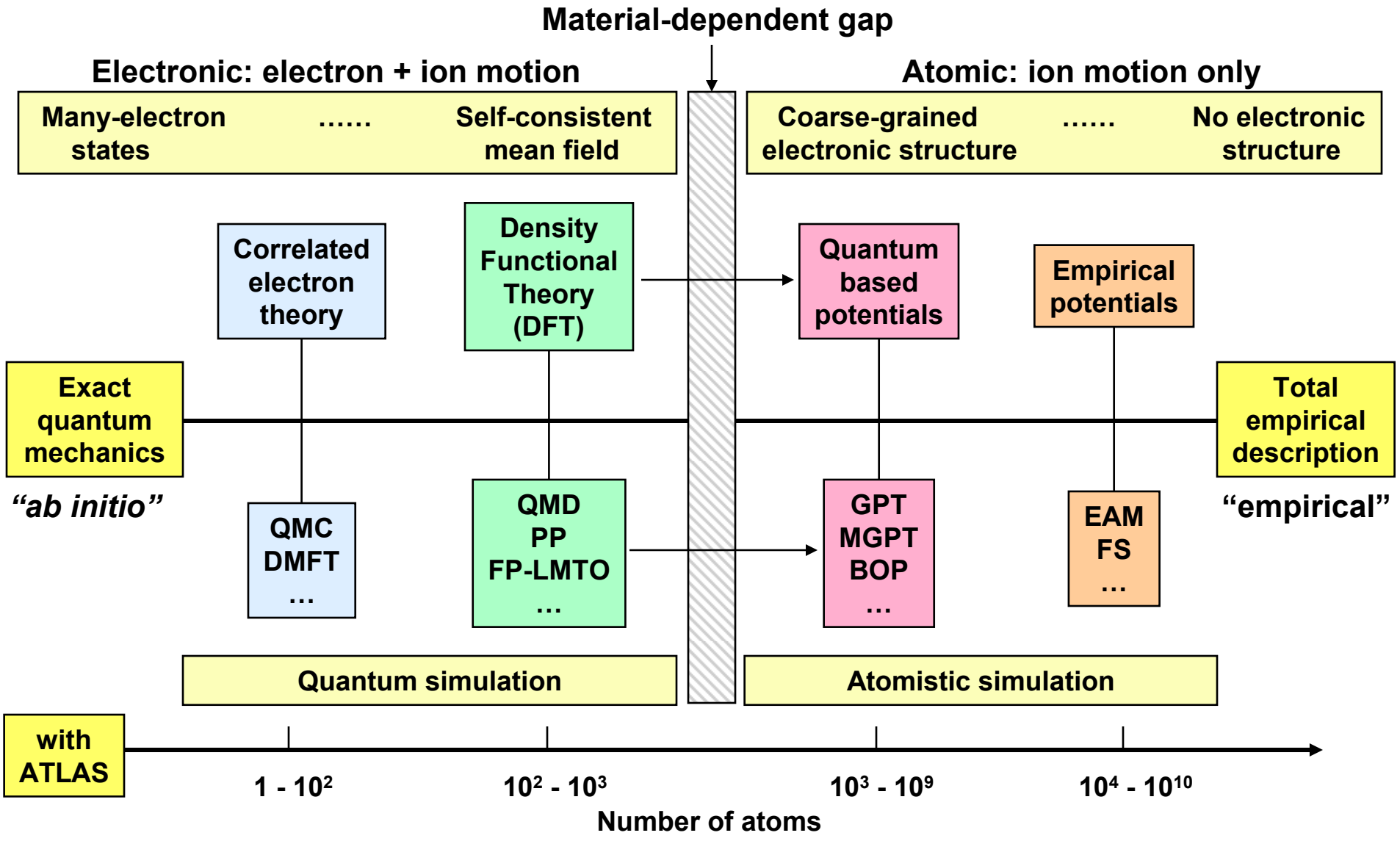
Simulation framework for SCC



Materials: *d*-electron transition metals and alloys, e.g., NiAl, at high temperatures

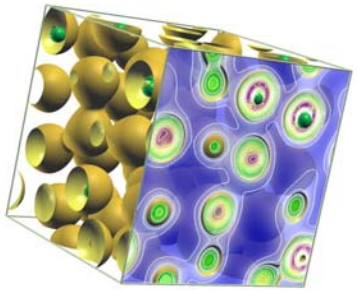
SciDAC focus: advanced simulation capability development

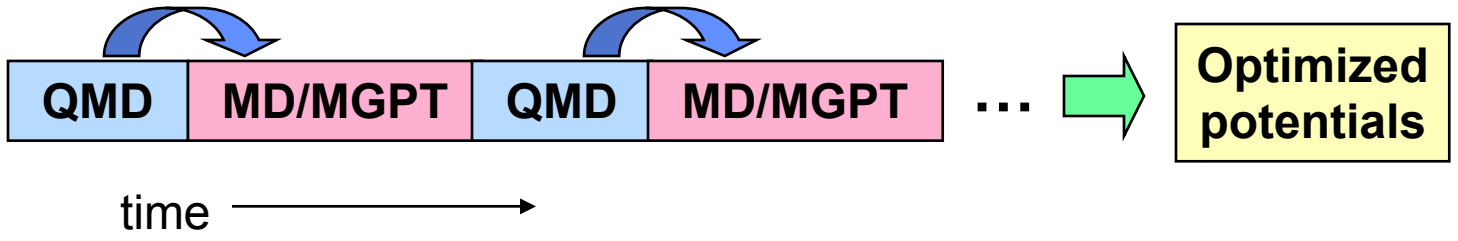
Bridging the gap from quantum mechanics to large scale atomistic simulation



Scientific challenge and strategy

- ***Scientific challenge***: bridge the length and time scale gap from quantum mechanics (~ 100 atoms) to ultra-scale atomistic simulation (billions of atoms) with quantum-level accuracy
- ***Strategy***: Direct coupling of *quantum simulations* (QMD) and MD simulations with quantum-based multi-ion potentials from *model generalized pseudopotential theory* (MGPT)
  - ATLAS enabling development of a robust hybrid QMD-MD/MGPT simulation capability for complex *d*-electron metals and alloys





  - Grand challenge goal: simulate 10^9 atoms at high temperature in prototype systems (e.g., Mo, NiAl) via MD/MGPT with accuracy of direct QMD simulation on 10^2 atoms

Scope of work and progress to date

1) **Development and testing of an efficient parallel QMD-MD/MGPT simulation code: *P^{3}MD***

   completed

2) **Development and application of robust simulation algorithm to optimize MGPT potentials**

   – first-generation algorithm established and applied to Mo prototype

   – excellent set of potentials obtained that accurately predict atomic structure of liquid Mo

   moving toward optimization

3) **Establishment of a QMD test data base and corresponding test simulations on derived MGPT potentials: *transferability***

   – extensive two-phase Mo melt simulations

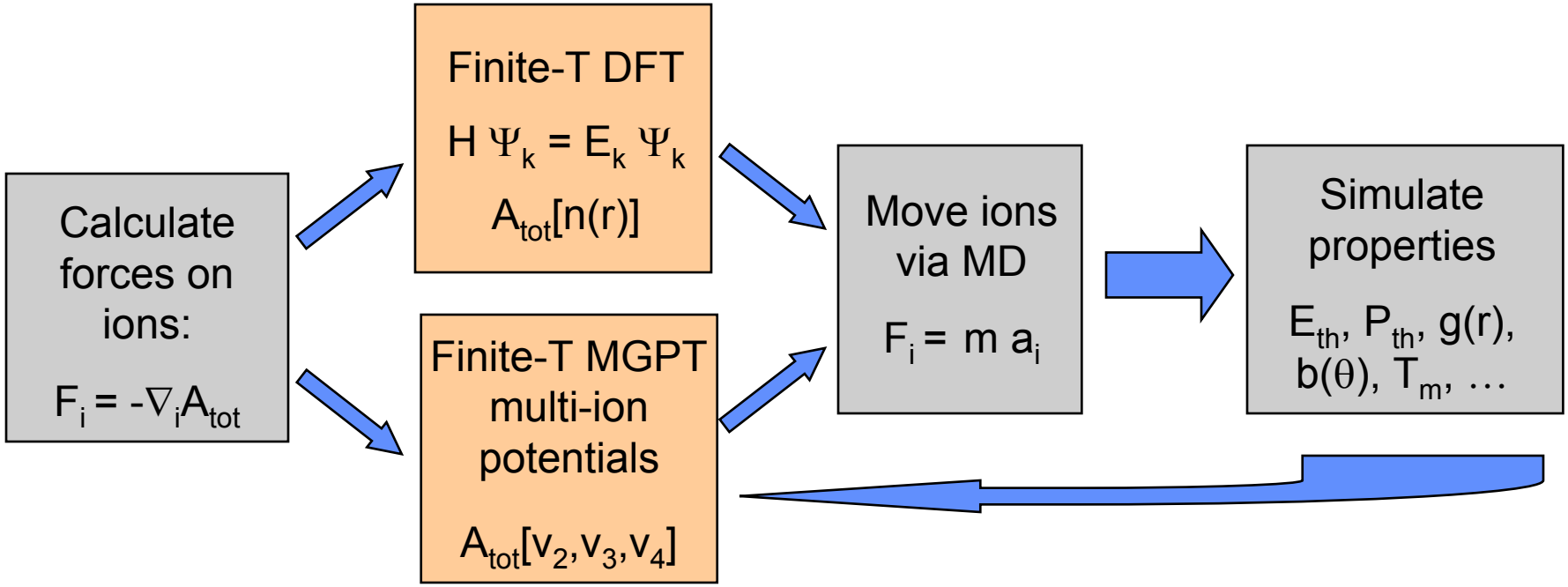
   in progress

4) **Billion-atom MD/MGPT demonstration simulations with optimized potentials**

   still to be completed

Hybrid QMD-MD/MGPT simulation code: *P³MD*

P³MD = Petascale, Plane-wave, Pseudopotential Molecular Dynamics



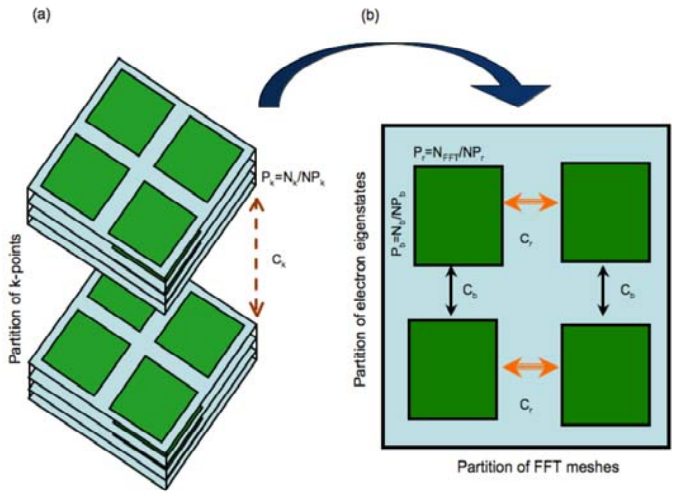
- **QMD mode: P³MD implements first-principles pseudopotential method**
  - plane wave basis for wavefunction expansion
  - treats 3-9 valence electrons per atom for *d*-electron transition metals
  - treats 50-250 transition-metal atoms with 1-10 k points
- **MD/MGPT mode: P³MD implements model generalized pseudopotential theory**

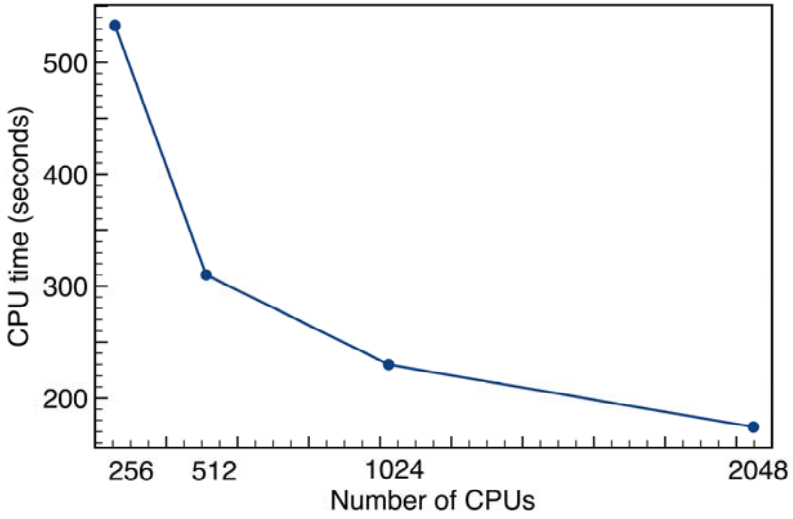
Efficient usage of HPC resources

Parallelization of *P³MD* code:

DFT forces: k points, energy bands, plane waves in reciprocal space spread across CPUs in optimal manner, with custom FFT used to move between real and reciprocal space

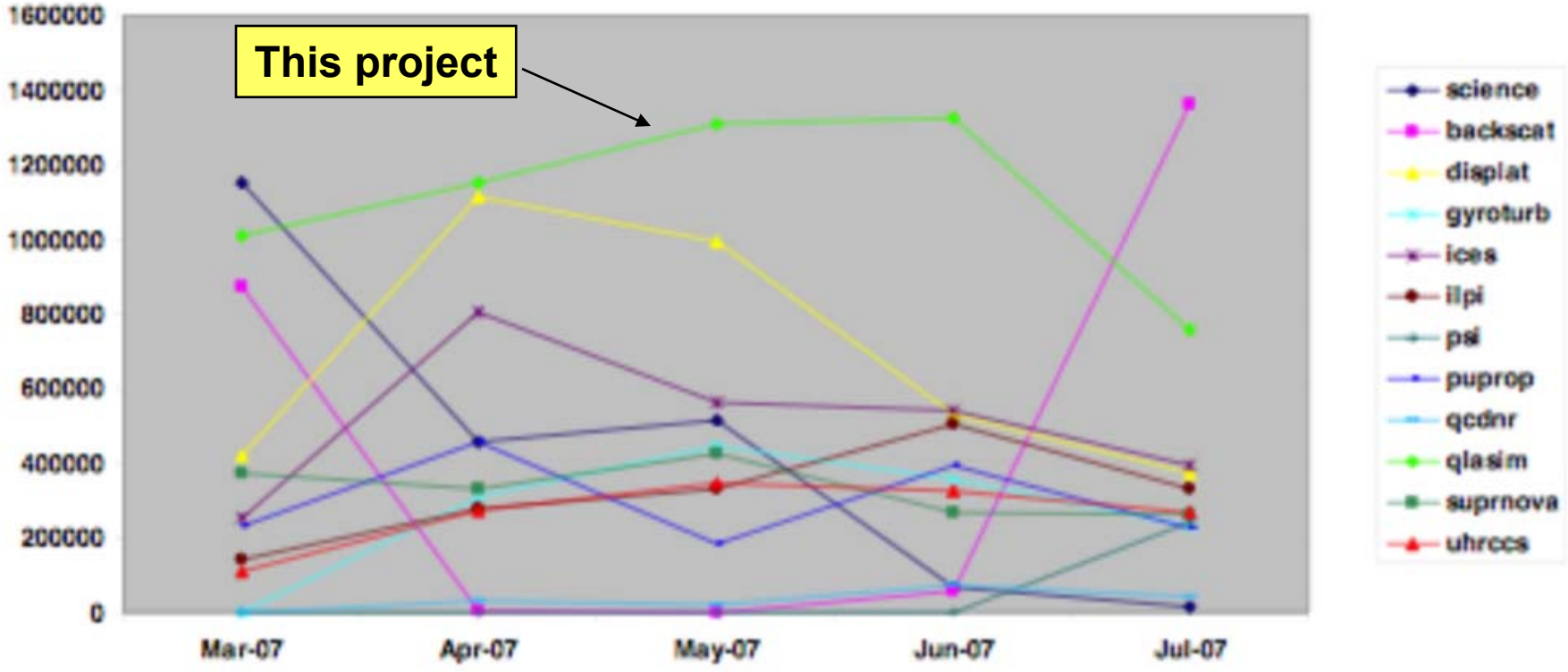
MD: performed on single CPU, assuming N is small (< 250)





Tests on QMD melt simulations show excellent efficiency

Atlas utilization on grand challenge projects



Average utilization by this project: 300,000 CPU/hours per week

Quantum-based MGPT multi-ion potentials

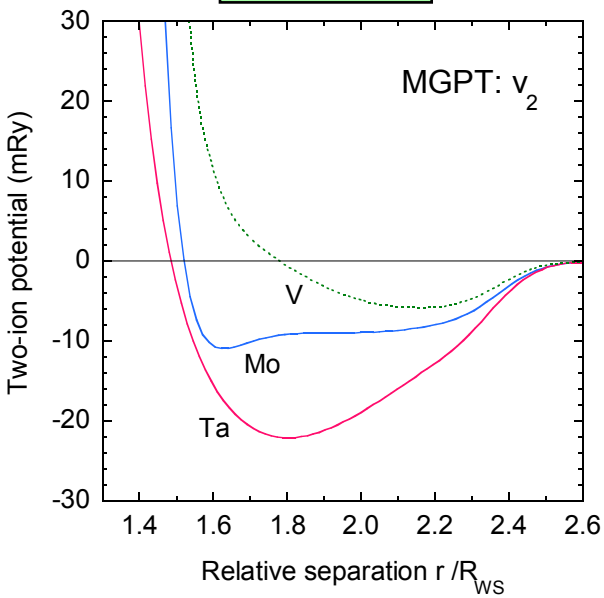
Total-energy expansion within DFT quantum mechanics

- MGPT potentials *derived* from systematic simplification of first-principles results
- standard analytic method: *canonical d bands* (universal form)
- fast matrix method: *non-canonical d bands* (materials specific form)

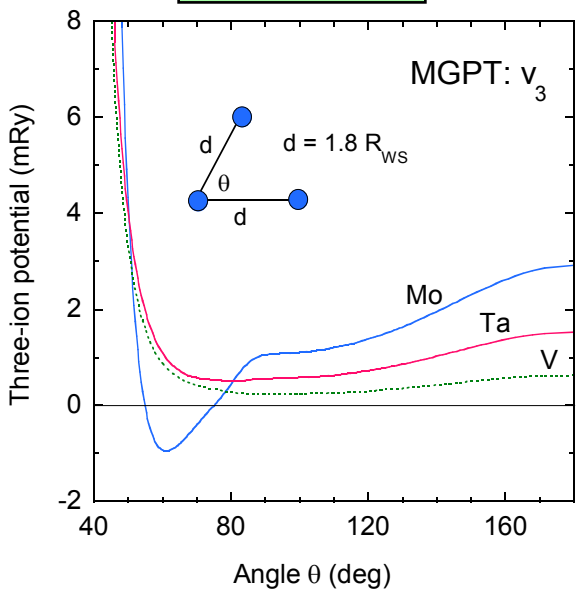
$$E_{tot}(R_1,\ldots,R_N) = NE_{vol}(\Omega) + \frac{1}{2}\sum_{i,j}{}' v_2(ij;\Omega) + \frac{1}{6}\sum_{i,j,k}{}' v_3(ijk;\Omega) + \frac{1}{24}\sum_{i,j,k,l}{}' v_4(ijkl;\Omega)$$

volume radial forces angular forces

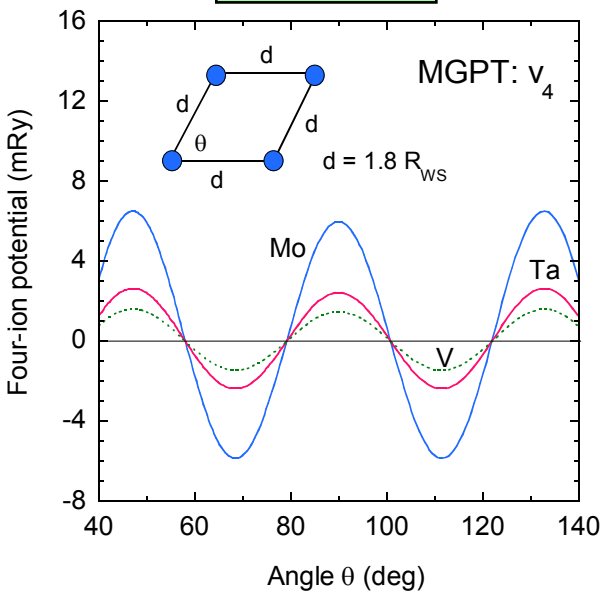
Two-ion



Three-ion



Four-ion



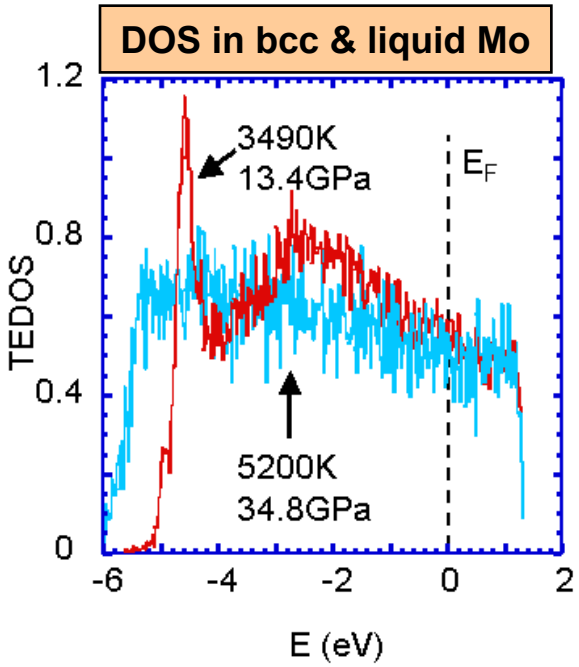
Importance of electron temperature T_{el}: three limits

(i) $T_{el} = 0$ and $T = T_{ion}$: low-temperature solid

- standard assumption in theory, even for high T
- adequate for most simple *sp*-bonded materials

(ii) $T_{el} = T_{ion} = T$: high-temperature solid and liquid

- important in *d*-electron transition metals due to to high, phase-dependent density of electronic states (DOS)
- also advantageous: allows one to leverage rich finite-T QMD data
- in MGPT, include through explicit T-dependent potentials:

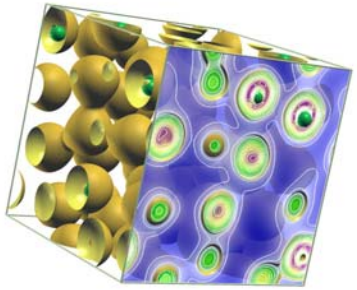


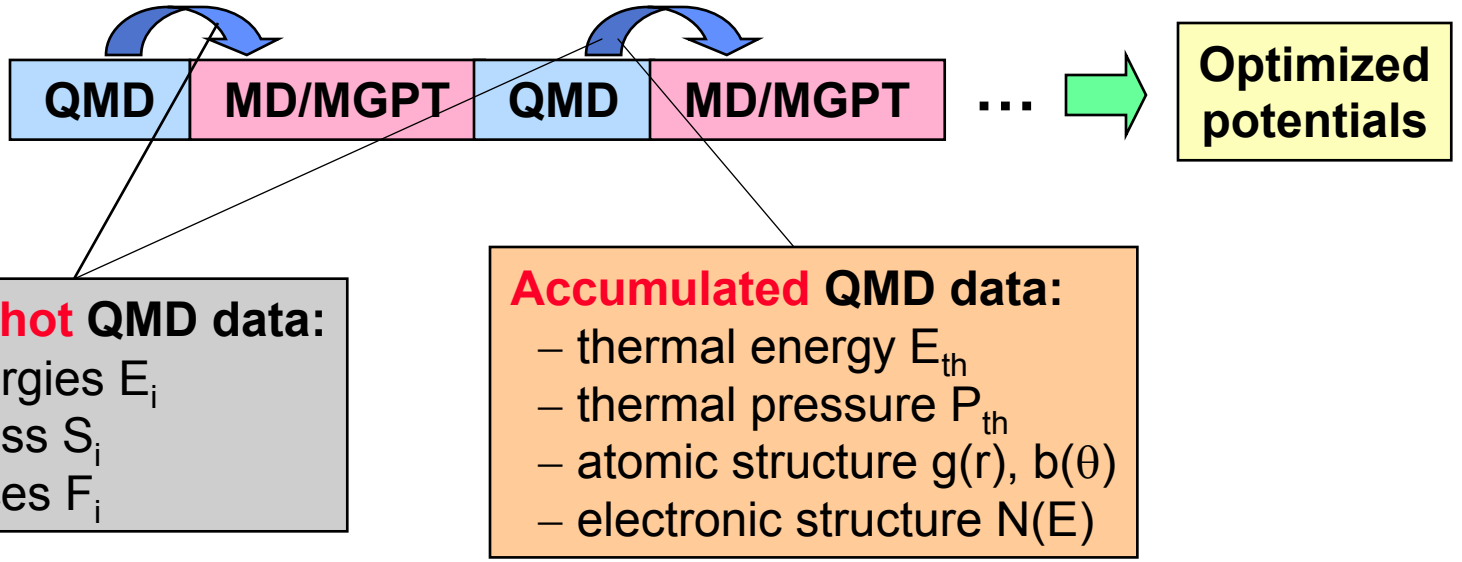
$$A_{tot}(R_1,\ldots,R_N) = NA_{vol}(\Omega,T) + \frac{1}{2}\sum_{i,j}{}' v_2(ij;\Omega,T) + \frac{1}{6}\sum_{i,j,k}{}' v_3(ijk;\Omega,T) + \frac{1}{24}\sum_{i,j,k,l}{}' v_4(ijkl;\Omega,T)$$

volume radial forces angular forces

(iii) $T_{el} >> T_{ion}$: ultra fast laser heating of solid or liquid

QMD-MD/MGPT simulation strategy





Optimization considerations:

(i) details of MGPT potentials functional form
- *canonical* vs *non-canonical d* bands
- finite-T systematic improvements

(ii) choice of constraining data
- snapshot vs accumulated QMD data
- additional static solid-phase data

Various options have been tried and a first-generation simulation scheme adopted

Adopted first-generation simulation scheme

MGPT potentials:

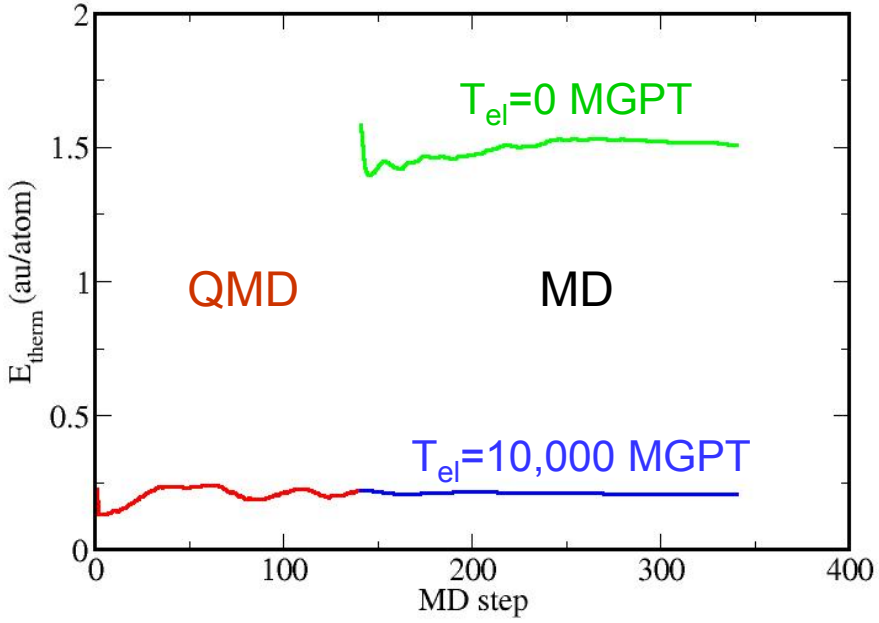
- *non-canonical d* bands
- explicit modeling of T_{el} contributions to A_{el}

Constraining data:

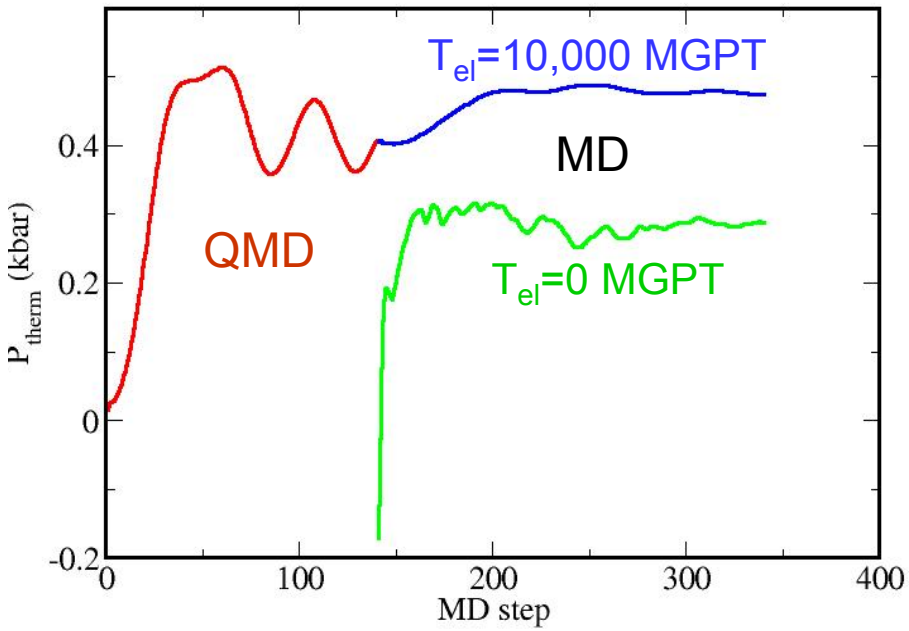
- blend of static bcc data at T_{el} + *snapshot* QMD energies in liquid from 20-30 configurations
- *predict* accumulated QMD properties as test

Smooth QMD → MD/MGPT transition and rapid convergence:

Instantaneous thermal energy



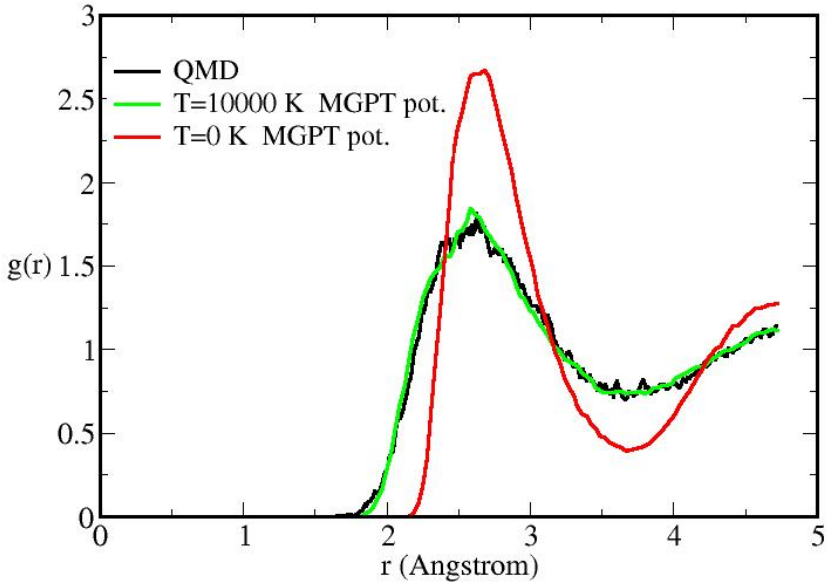
Instantaneous thermal pressure



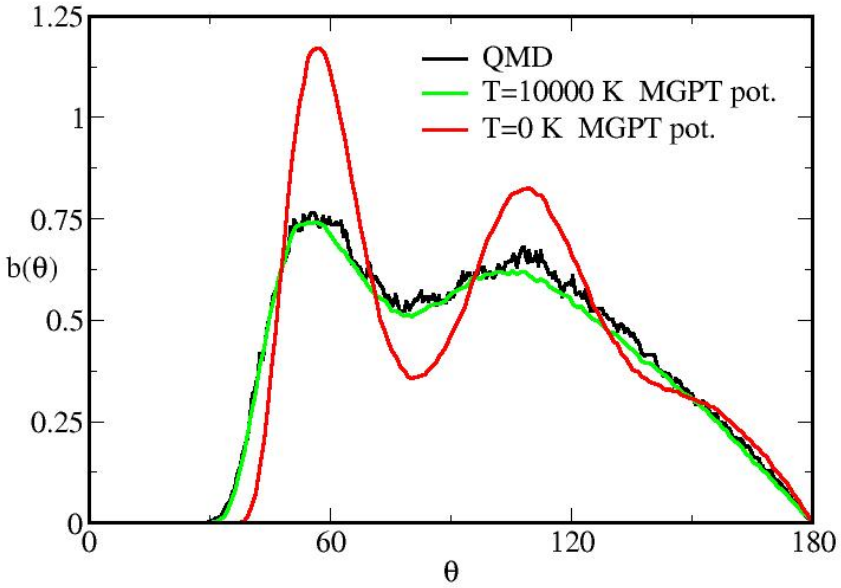
Simulation conditions: N = 54 atoms, T_{ion}= 10,000 K, Ω = 105.1 au

First-generation Mo potentials accurately predict atomic structure of liquid

Radial distribution function g(r)



Bond-angle distribution function b(θ)

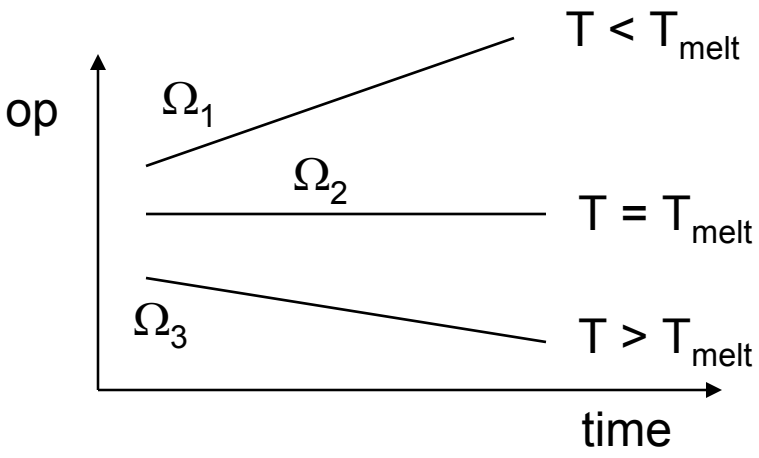


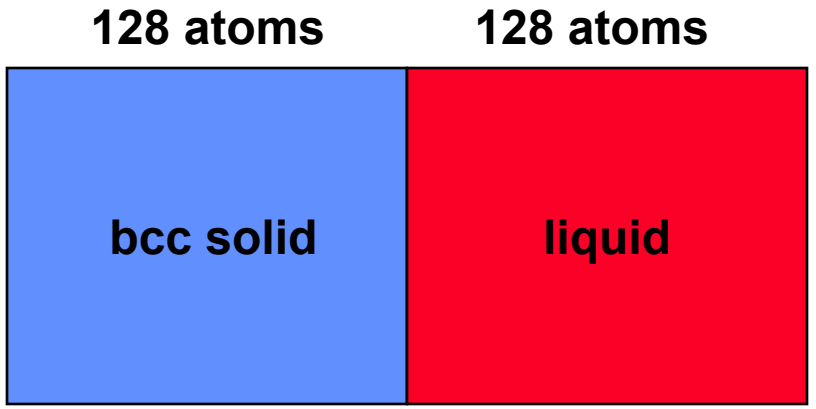
Simulation conditions: N = 54 atoms, T_{ion}= 10,000 K, Ω = 105.1 au

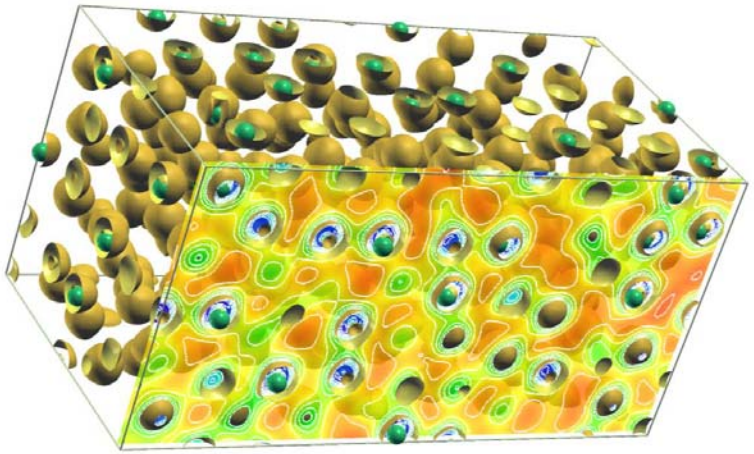
Similar results have been obtained over a wide range of volumes at both 5000 and 10,000 K

Two-phase baseline QMD melt simulation in Mo

- **Parameters of simulation**
  - N = 256 atoms (minimum size)
  - constant T, constant NΩ
  - 1-2 ps runs
  - 1 k point
- **Diagnostic strategy**
  - monitor order parameter (op) rather than interface position
  - for fixed T, do runs at several volumes of interest

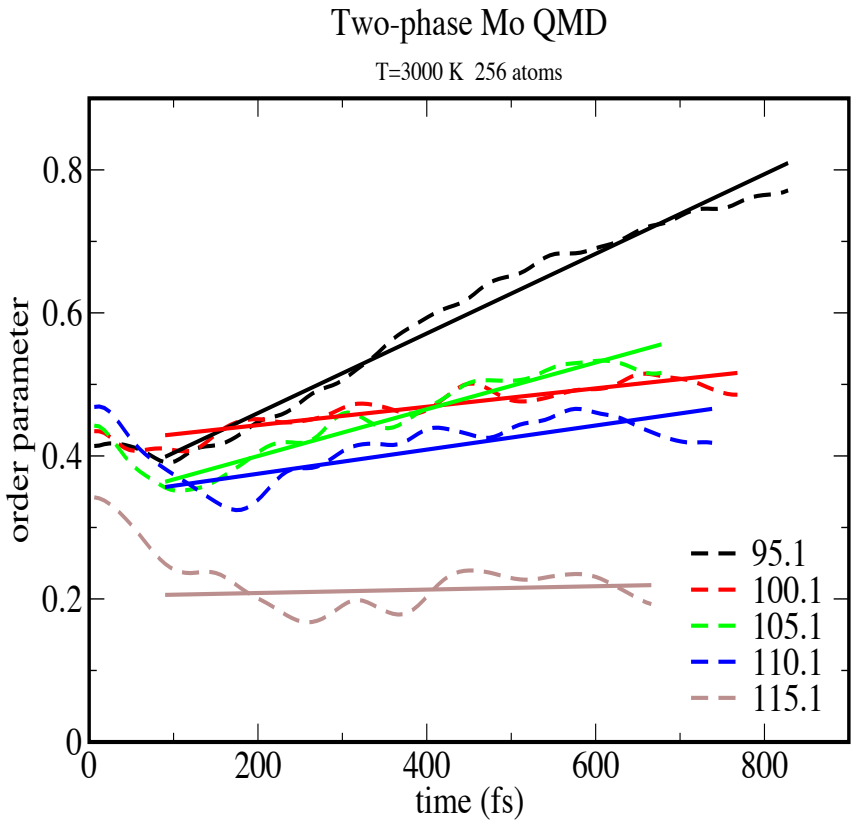


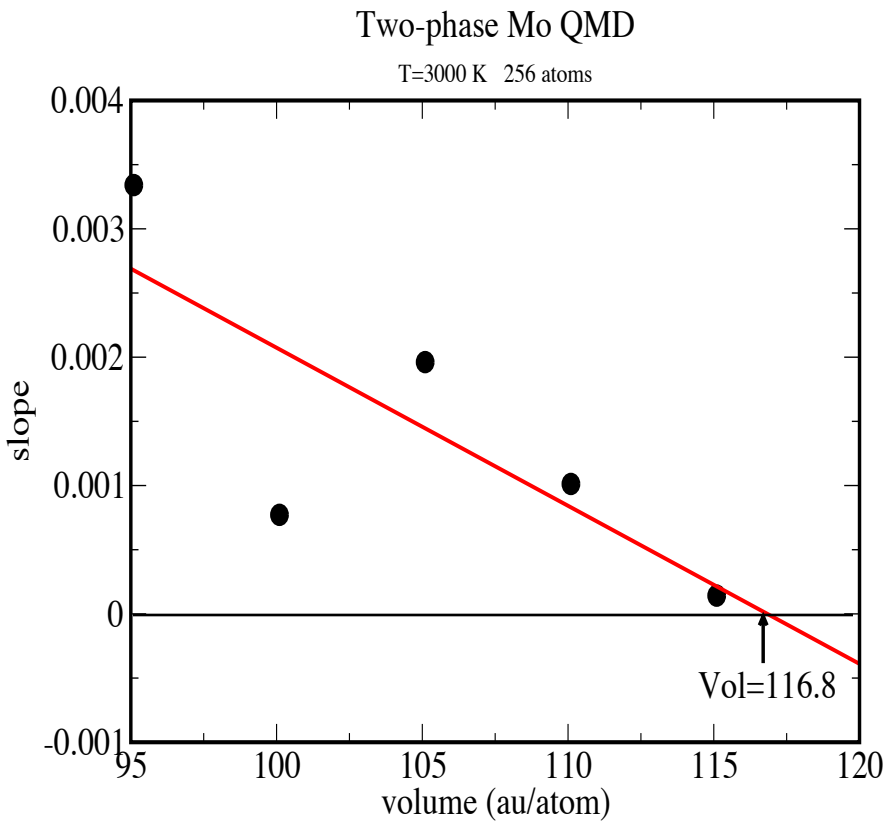




Snapshot of Mo QMD melt:
T = 3500 K and P = 35 GPa

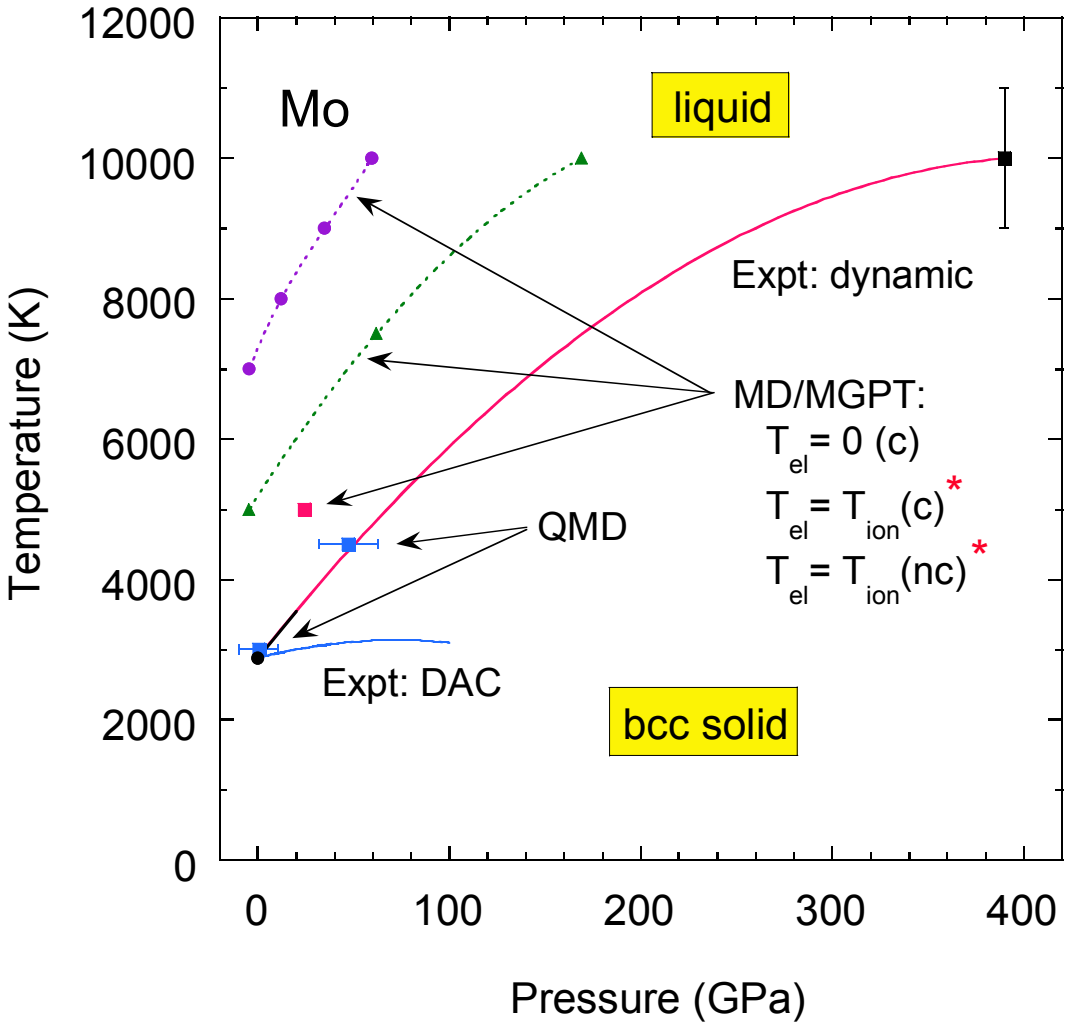
Two-phase QMD melt analysis: an example





Same methodology applies to MD/MGPT melt simulations

Mo melt curve: sensitive test of MGPT potentials



For MD/MGPT results:

(c) = *canonical* d bands

(nc) = *non-canonical* d bands

*early forms of T-dependent MGPT potentials

Additional simulated melt points from QMD and MD/MGPT with current potentials are in progress

Conclusions and expected scientific impact

- **A new hybrid QMD-MD/MGPT simulation algorithm has been developed to obtain *quantum-level accurate* interatomic potentials**
  - excellent first-generation MGPT potentials have been developed in a Mo prototype
  - in the coming year, we expect to complete algorithm optimization, potential transferability testing, and initial large scale applications

If we are successful …

- **New predictive science tool for materials extending continuously from *nano*meters to *micro*meters**
  - essential to SciDAC goal of first-principles SCC multiscale modeling
  - permit fundamental investigations beyond current capabilities
- **Will enable accurate advanced programmatic applications**
  - non-equilibrium multiphase equations of state with phase kinetics
  - dynamic strength and failure modeling up to micron length scales